

Attorney's Docket No. SAE-0039

## ORIGINAL CLAMS

International Application No.: PCT/JP2004/013392

Applicant: Kazuko Matsumoto et al.

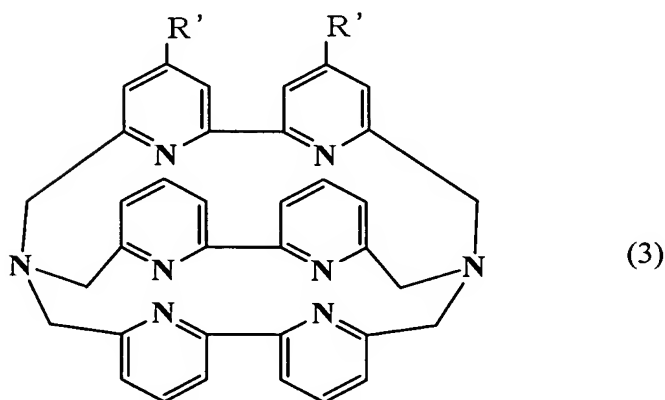
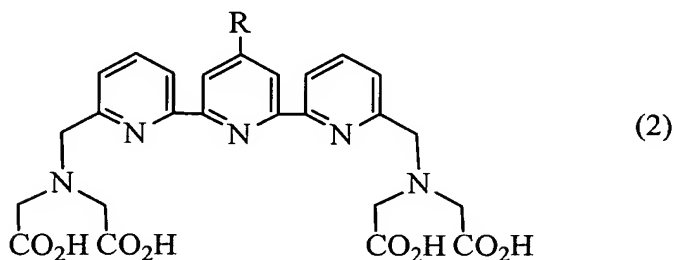
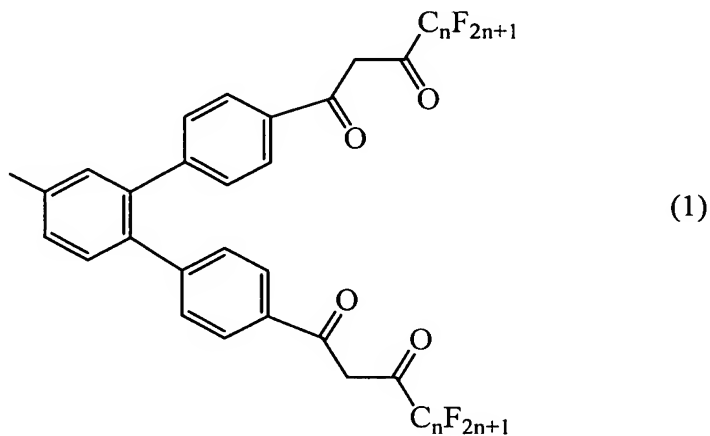
Title: NOVEL FINE FLUORESCENT PARTICLE

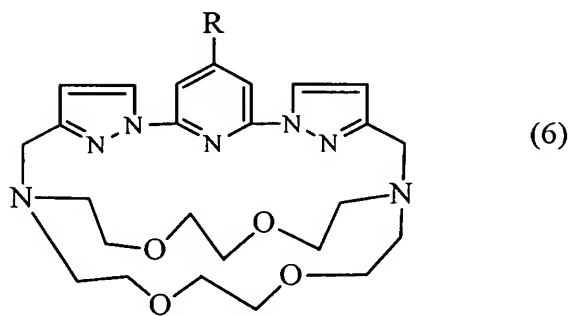
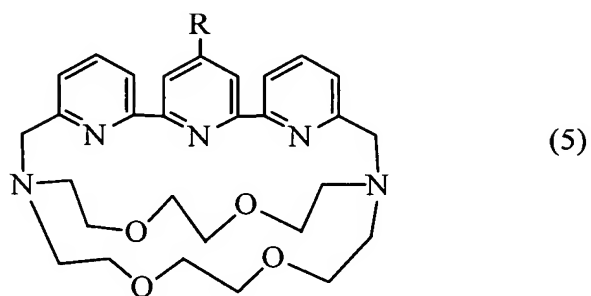
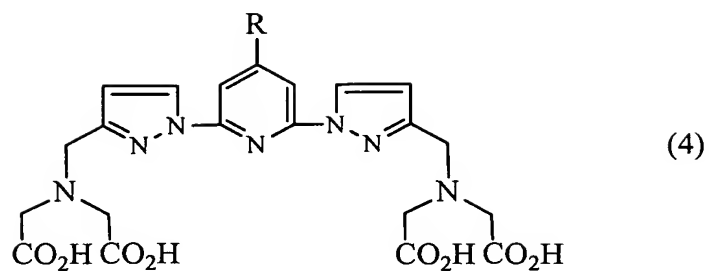
Rader, Fishman & Grauer PLLC

**A**

# CLAIMS

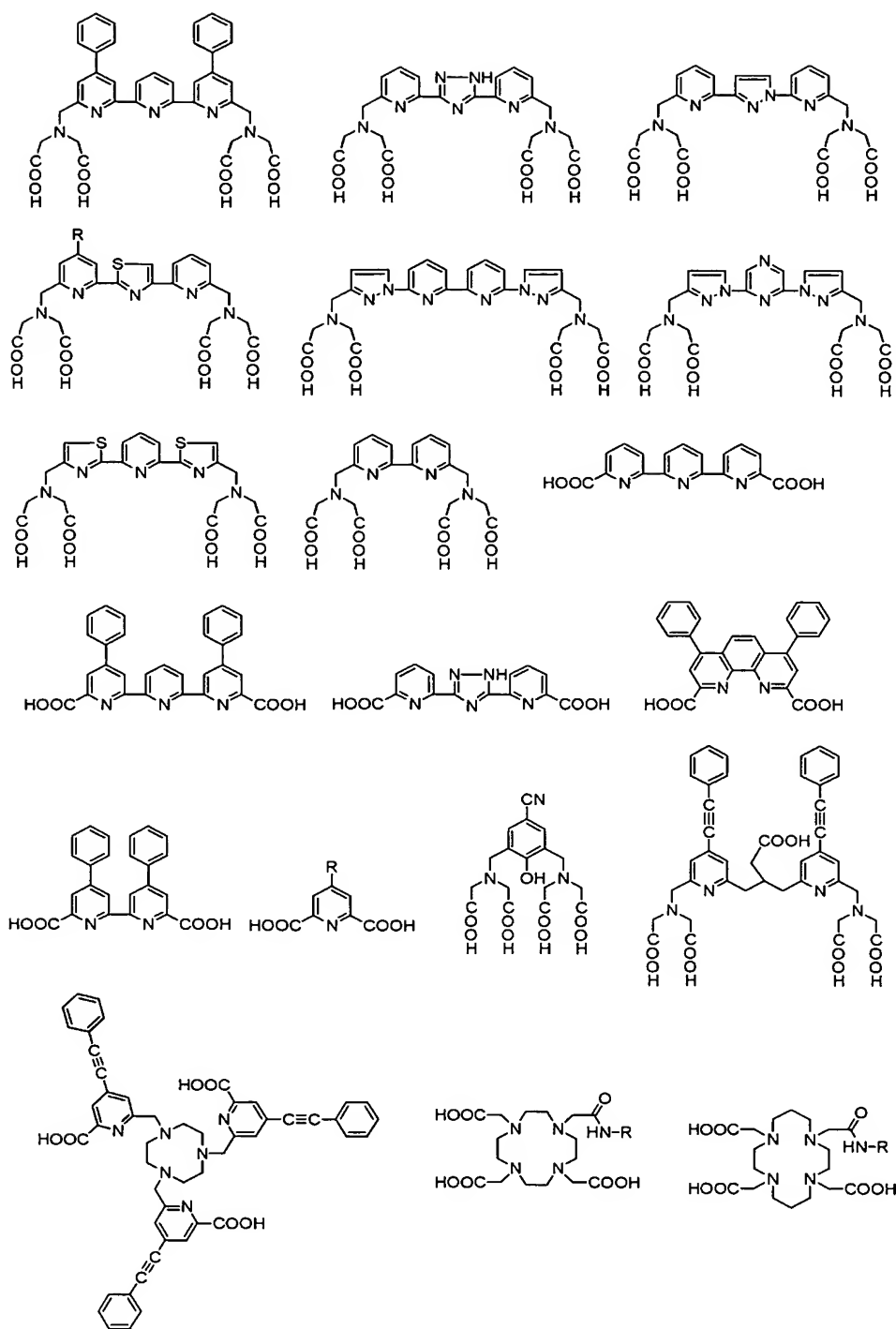
1. Silica particles containing a fluorescent rare-earth complex comprising a fluorescent rare-earth complex.
2. The silica particles according to claim 1, wherein the fluorescent rare-earth complex comprises at least one kind of a compound selected from the group consisting of the compounds represented by the following formulae (1) to (6)





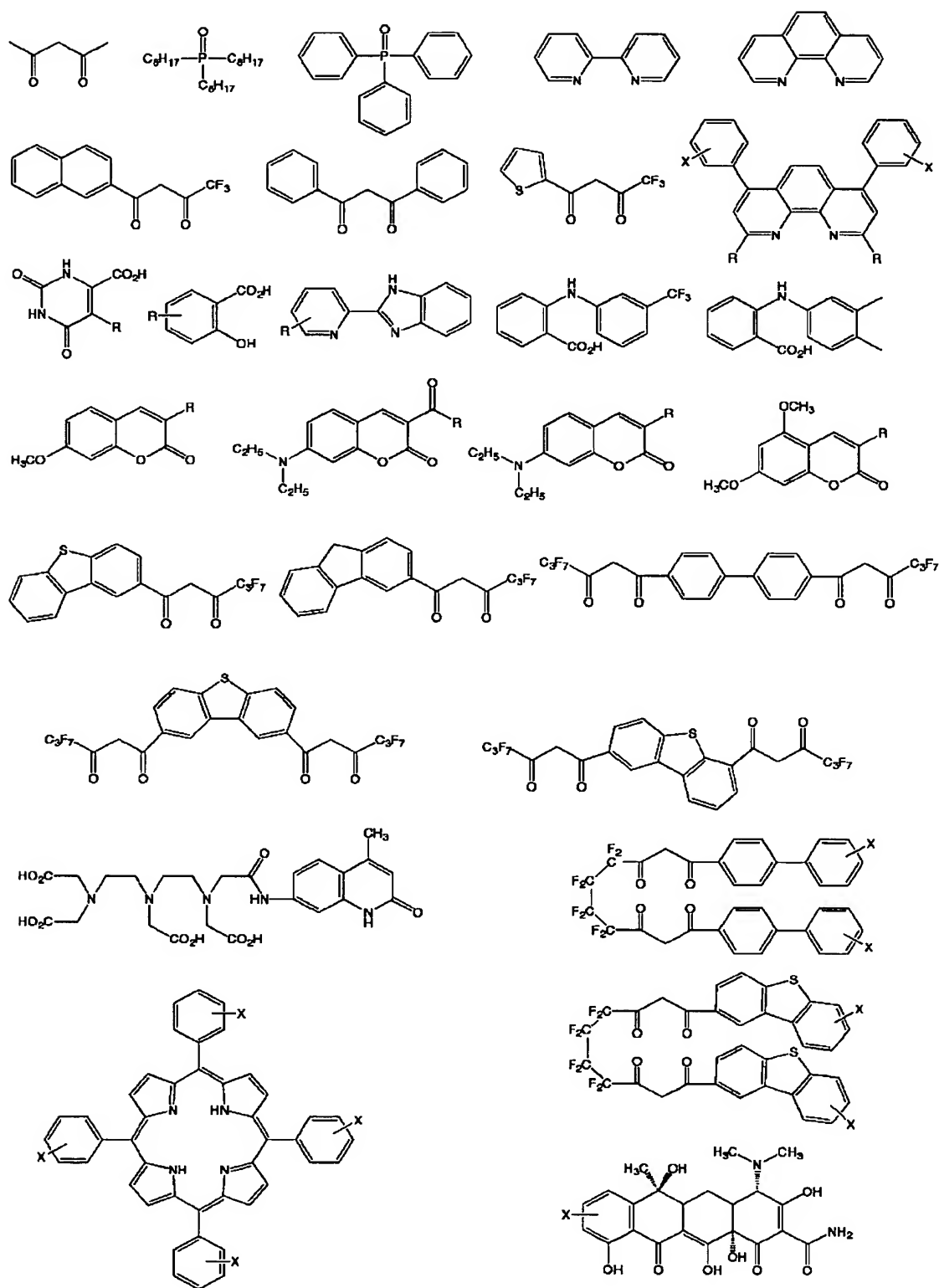
as a ligand.

3. The silica particles according to claim 1, wherein the fluorescent rare-earth complex comprising at least one kind of a compound selected from the group consisting of the compounds represented by the following formulae



as a ligand.

4. The silica particles according to claim 1, wherein the fluorescent rare-earth complex comprising at least one kind of a compound selected from the group consisting of the compounds represented by the following formulae



as a ligand.

5. The silica particles according to any one of claims 1 to 4, wherein the fluorescent rare-earth complex is a complex of terbium or europium.
6. The silica particles according to any one of claims 1 to 5, wherein the silica particles containing a fluorescent rare-earth complex is produced by an emulsion polymerization method.
7. The silica particles according to any one of claims 1 to 6, wherein silica particles selectively contain the fluorescent rare-earth complex mainly in an inner layer thereof.
8. The silica particles according to any one of claims 1 to 7, wherein the bonding groups to a substance to be labeled is introduced to the silica particles containing the fluorescent rare-earth complex.
9. The silica particles according to claim 8, wherein the bonding groups are cyano groups.
10. The silica particles according to any one of claims 1 to 9, wherein intercalator to double-stranded DNA is introduced to the silica particles containing the fluorescent rare-earth complex.
11. The silica particles according to any one of claims 1 to 9, wherein intercalator to double-stranded RNA is introduced to the silica particles containing the fluorescent rare-earth complex.
12. The silica particles according to any one of claims 1 to 9, wherein intercalator to double-stranded PNA is introduced to the silica particles containing the fluorescent rare-earth complex.
13. The silica particles according to any one of claims 1 to 9, wherein intercalator to double-stranded DNA/RNA hybrid is introduced to the silica particles containing the fluorescent rare-earth complex.
14. The silica particles according to any one of claims 1 to 9, wherein intercalator to double-stranded DNA/PNA hybrid is introduced to the silica particles containing the fluorescent rare-earth complex.
15. The silica particles according to any one of claims 1 to 9, wherein intercalator to double-stranded RNA/PNA hybrid is introduced to the silica particles containing the fluorescent rare-earth complex.
16. The silica particles according to any one of claims 10 to 15, wherein intercalator is monovalent.

17. A fluorescent labeling agent comprising the silica particles according to any one of claims 1 to 16.
18. A fluorescence labeled nucleic acid probe, wherein the nucleic acid probe is bonded at the surface of the silica particles according to any one of claims 1 to 16.
19. The nucleic acid probe according to claim 18, wherein the nucleic acid is DNA.
20. The nucleic acid probe according to claim 18, wherein the nucleic acid is RNA.
21. The nucleic acid probe according to claim 18, wherein the nucleic acid is PNA.
22. A method for detection of double-stranded DNA using the silica particles containing the fluorescent rare-earth complex, which is introduced with the intercalator to the double-stranded DNA according to claim 10.
23. A method for detection of double-stranded RNA using silica particles containing the fluorescent rare-earth complex, which is introduced with the intercalator to the double-stranded RNA according to claim 11.
24. A method for detection of double-stranded PNA using the silica particles containing the fluorescent rare-earth complex, which is introduced with the intercalator to the double-stranded PNA according to claim 12.
25. A method for detection of a double-stranded DNA/RNA hybrid using the silica particles containing the fluorescent rare-earth complex, which is introduced with the intercalator to the double-stranded DNA/RNA hybrid according to claim 13.
26. A method for detection of a double-stranded DNA/PNA hybrid using the silica particles containing the fluorescent rare-earth complex, which is introduced with the intercalator to the double-stranded DNA/PNA hybrid according to claim 14.
27. A method for detection of a double-stranded RNA/PNA hybrid using the silica particles containing the fluorescent rare-earth complex, which is introduced with the intercalator to the double-stranded RNA/PNA hybrid according to claim 15.
28. A kit for target molecule determination comprising a molecular species containing the fluorescent labeling agent according to claim 17, or markers containing said fluorescent labeling agent, and material for target molecule determination.

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# **AMENDMENT**

(Amended Claims under PCT Article 34)

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Title: NOVEL FINE FLUORESCENT PARTICLE

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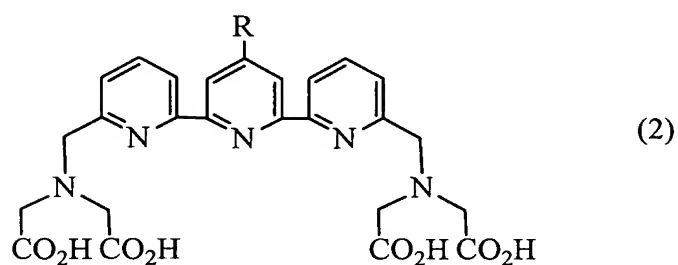
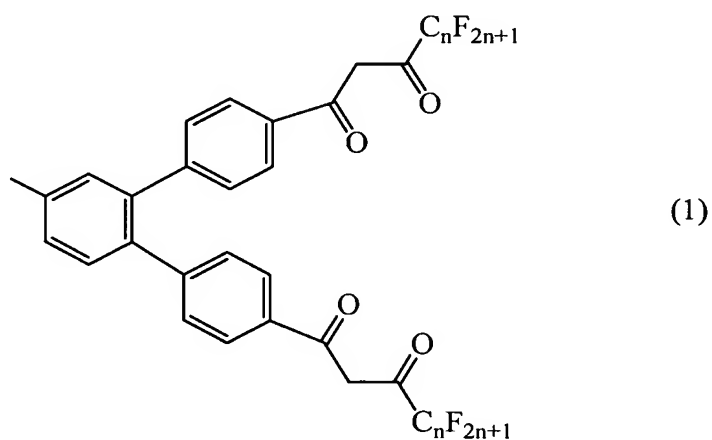
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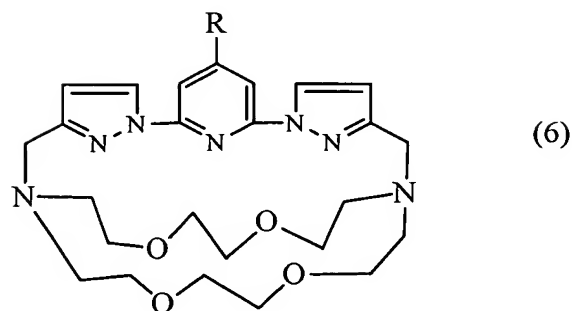
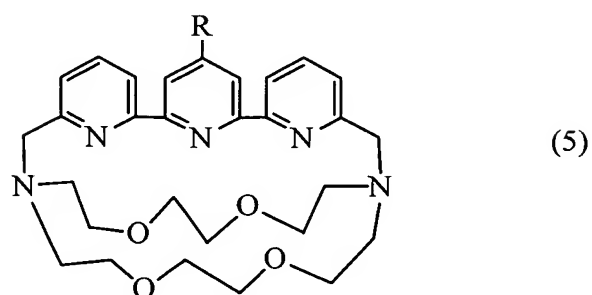
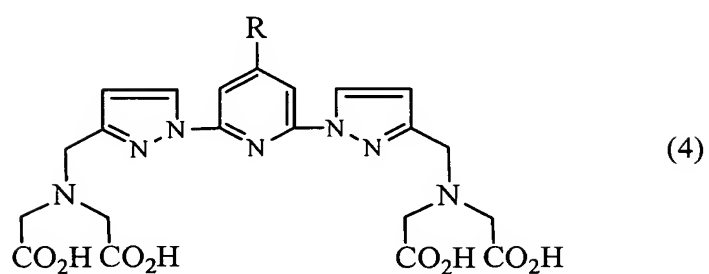
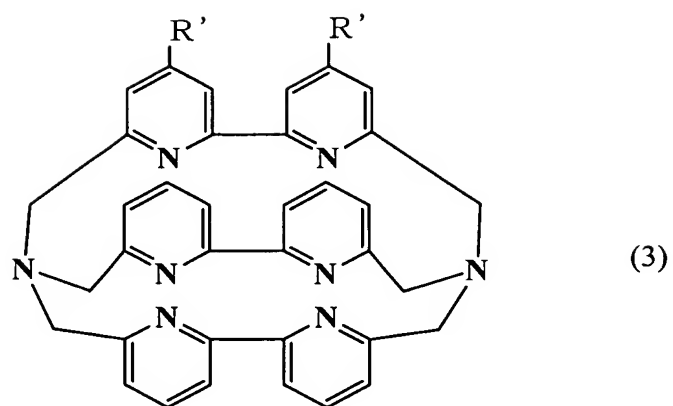


# Claims

(Amended under PCT Article 34)

1. (Amended) Silica particles containing a fluorescent rare-earth complex selectively in an inner layer thereof and the surface of which substantially comprises silica.
2. The silica particles according to claim 1, wherein the fluorescent rare-earth complex comprises at least one kind of a compound selected from the group consisting of the compounds represented by the following formulae (1) to (6)

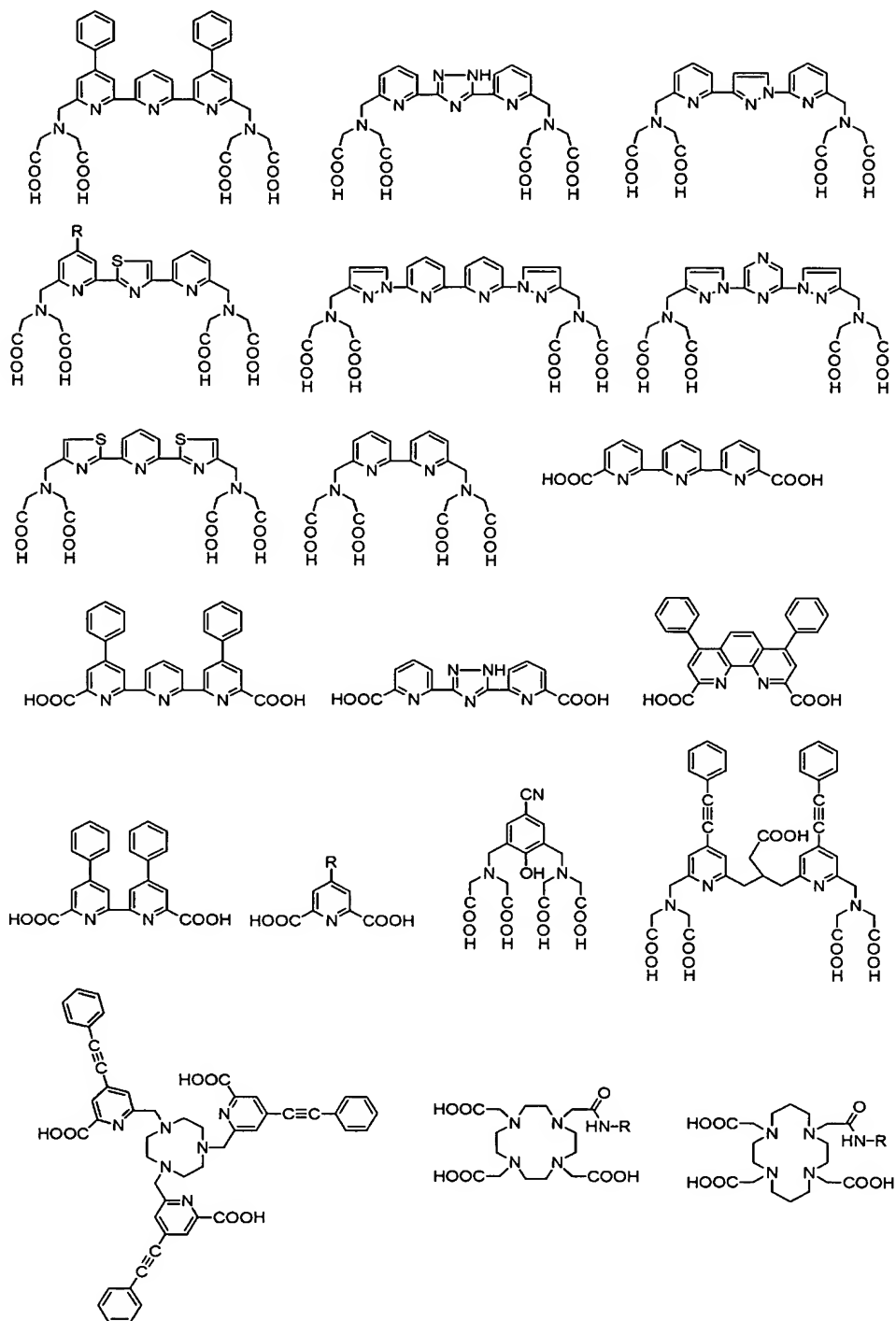




as a ligand.

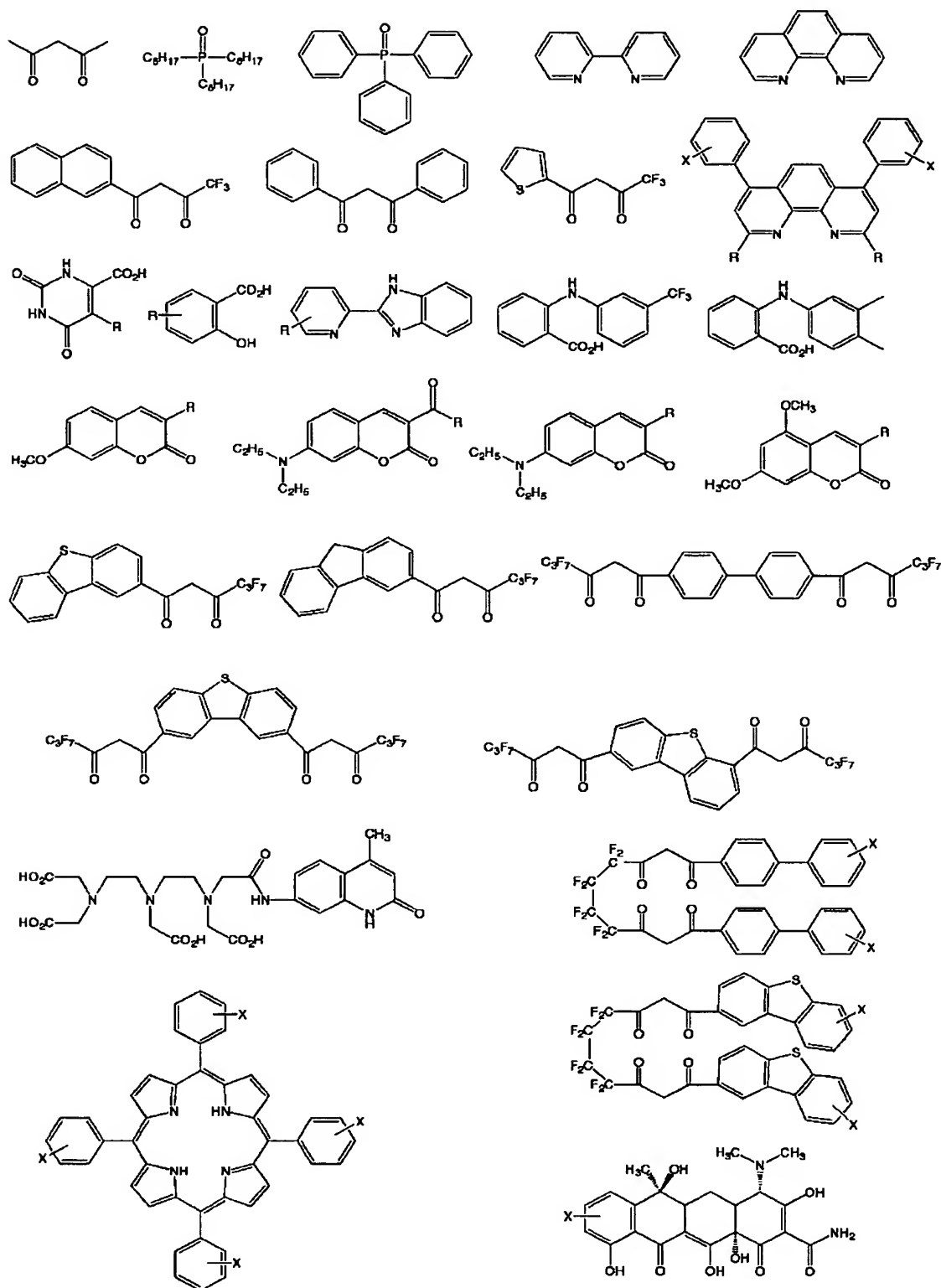
3. The silica particles according to claim 1, wherein the fluorescent rare-earth complex comprising at least one kind of a compound selected from the group consisting of the compounds

represented by the following formulae



as a ligand.

4. The silica particles according to claim 1, wherein the fluorescent rare-earth complex comprising at least one kind of a compound selected from the group consisting of the compounds represented by the following formulae



as a ligand.

5. The silica particles according to any one of claims 1 to 4, wherein the fluorescent rare-earth complex is a complex of terbium or europium.

6. The silica particles according to any one of claims 1 to 5, wherein the silica particles containing a fluorescent rare-earth complex is produced by an emulsion polymerization method.
7. (Deleted)
8. The silica particles according to any one of claims 1 to 7, wherein the bonding groups to a substance to be labeled is introduced to the silica particles containing the fluorescent rare-earth complex.
9. The silica particles according to claim 8, wherein the bonding groups are cyano groups.
10. The silica particles according to any one of claims 1 to 9, wherein intercalator to double-stranded DNA is introduced to the silica particles containing the fluorescent rare-earth complex.
11. The silica particles according to any one of claims 1 to 9, wherein intercalator to double-stranded RNA is introduced to the silica particles containing the fluorescent rare-earth complex.
12. The silica particles according to any one of claims 1 to 9, wherein intercalator to double-stranded PNA is introduced to the silica particles containing the fluorescent rare-earth complex.
13. The silica particles according to any one of claims 1 to 9, wherein intercalator to double-stranded DNA/RNA hybrid is introduced to the silica particles containing the fluorescent rare-earth complex.
14. The silica particles according to any one of claims 1 to 9, wherein intercalator to double-stranded DNA/PNA hybrid is introduced to the silica particles containing the fluorescent rare-earth complex.
15. The silica particles according to any one of claims 1 to 9, wherein intercalator to double-stranded RNA/PNA hybrid is introduced to the silica particles containing the fluorescent rare-earth complex.
16. The silica particles according to any one of claims 10 to 15, wherein intercalator is monovalent.
17. A fluorescent labeling agent comprising the silica particles according to any one of claims 1 to 16.
18. A fluorescence labeled nucleic acid probe, wherein the nucleic acid probe is bonded at

the surface of the silica particles according to any one of claims 1 to 16.

19. The nucleic acid probe according to claim 18, wherein the nucleic acid is DNA.
20. The nucleic acid probe according to claim 18, wherein the nucleic acid is RNA.
21. The nucleic acid probe according to claim 18, wherein the nucleic acid is PNA.
22. A method for detection of double-stranded DNA using the silica particles containing the fluorescent rare-earth complex, which is introduced with the intercalator to the double-stranded DNA according to claim 10.
23. A method for detection of double-stranded RNA using silica particles containing the fluorescent rare-earth complex, which is introduced with the intercalator to the double-stranded RNA according to claim 11.
24. A method for detection of double-stranded PNA using the silica particles containing the fluorescent rare-earth complex, which is introduced with the intercalator to the double-stranded PNA according to claim 12.
25. A method for detection of a double-stranded DNA/RNA hybrid using the silica particles containing the fluorescent rare-earth complex, which is introduced with the intercalator to the double-stranded DNA/RNA hybrid according to claim 13.
26. A method for detection of a double-stranded DNA/PNA hybrid using the silica particles containing the fluorescent rare-earth complex, which is introduced with the intercalator to the double-stranded DNA/PNA hybrid according to claim 14.
27. A method for detection of a double-stranded RNA/PNA hybrid using the silica particles containing the fluorescent rare-earth complex, which is introduced with the intercalator to the double-stranded RNA/PNA hybrid according to claim 15.
28. A kit for target molecule determination comprising a molecular species containing the fluorescent labeling agent according to claim 17, or markers containing said fluorescent labeling agent, and material for target molecule determination.